ARKIV FÖR FYSIK Band 17 nr 17

Communicated 14 October 1959 by M. SIEGBAHN and E. HULTHÉN

The decay of Tl^{201}

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With 8 figures in the text

ABSTRACT

Using beta- and gamma-spectroscopic techniques, the decay of Tl^{201} has been investigated. The half-life of Tl^{201} was remeasured to be (73.5 ± 0.8) h. From internal conversion measurements in a double-focusing beta spectrometer, the energies of the four transitions previously known were found to be 30.60, 32.19, 135.34 and 167.43 keV. In addition, conversion lines of a new, weak transition of energy 165.88 keV were detected. From the measured conversion ratios the multipolarities of all the transitions were proved to be $M1$, although, for the 165.88 keV transition, an $E2$ admixture of up to 7 % cannot be excluded. A further proof of the multipolarity assignments of the 135.34 and 167.43 keV transitions was obtained from a determination of absolute internal conversion coefficients using the method of external conversion. Electron-gamma and gamma-gamma coincidence measurements were performed. The collected experimental information is in agreement with the following level sequence: $3/2-$(0 keV), $5/2-$(1.57 keV), $3/2-$(32.19 keV), and $3/2-$(167.49 keV); *or* $3/2-$(0 keV), $1/2-$(1.57 keV), $1/2-$ or $3/2-$(32.19 keV), and $1/2-$ or $3/2-$(167.49 keV). Electron-capture branching ratios are given.

The investigation also gives information about relative intensities of the KLL Auger lines in mercury, and relative photo cross-sections for L subshells of uranium, $(\tau_{L_{I}}+\tau_{L_{II}})/\tau_{L_{III}}$. The experimental results are in both cases compared with theoretical calculations.

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Introduction

Tl^{201} was first discovered by Neumann and Perlman [1] in the decay chain $Bi^{201} \rightarrow Pb^{201} \rightarrow Tl^{201} \rightarrow Hg^{201}$. They determined its half-life to be 72 ± 3 hours. In a beta-spectroscopic study Bergström *et al.* [2] found gamma rays of energies 30.5, 32.1, 135.3 and 167.6 keV belonging to the decay of Tl^{201}. They were all classified as $M1$ transitions from L-subshell ratios obtained from photographic films. From the failure to observe a gamma ray of 0.55 MeV, known to be emitted in the beta decay of Au^{201} [3], the total decay energy of Tl^{201} was supposed to be less than 0.55 MeV. Bergkvist *et al.* [4] found energies and multipolarity assignments in agreement with those above. They also performed electron-gamma coincidence measurements and found that the 135 keV transition is in coincidence with each of the two low-energy transitions, whereas no coincidences were observed between the 167 keV transition and any other transition. Assuming an $s_{1/2}$ ground state in Tl^{201}, the following level assignment for Hg^{201} was found to agree with their experiments: $p_{3/2}$(0 keV), $5/2-$(1.5 keV), $3/2-$(32.1 keV), and $1/2-$(167.2 keV).

Although these preliminary measurements give a good deal of information about the decay of Tl^{201}, it must be concluded that there are still many uncertain features in the construction of the level scheme. A more detailed investigation was therefore considered called for, with emphasis on a search for new gamma rays and on more precise determinations of intensities and energies of the transitions. It was hoped that the result of such measurements would throw more light on the level scheme of Hg^{201} in particular and on the level systematics of the odd Hg isotopes in general.

A. Experimental procedures and results

1. Source preparation

The Tl^{201} activity was obtained as a daughter product of Pb^{201} which was produced by bombarding thallium metal with about 40 MeV protons in the synchrocyclotron of the Gustaf Werner Institute in Uppsala. The chemical method used for separating the active lead from the irradiated thallium has been described elsewhere [5]. Then, in most cases, the grown-in thallium activity was, after about 18 hours, extracted with ether saturated with 6 N HCl and evaporated onto an aluminium foil (about 2.5 mg/cm^2) using a special vacuum evaporation device [6]. The aluminium foil was used as backing of the beta-spectrometer sources, which had a size of 2×12 mm^2. Since several lead isotopes were produced in the cyclotron bombardment, the sources contained also activities of the neighbouring isotopes, mainly Tl^{200} and Tl^{202}.

For some special investigations of the conversion-line spectrum, thinner and purer sources had to be used. These sources were prepared by electromagnetic isotope separation of the active lead isotopes (bombarding energy about 60 MeV in this case). The Pb^{201} activity was collected directly onto the aluminium backing foil and the measurements were carried out on the grown-in Tl^{201} activity.

2. *Half-life measurement*

The decay of an isotope-separated sample was followed in an end-window G.M. counter for about one month. In order to avoid errors due to small changes in the efficiency of the experimental set-up, the counting rate of the Tl^{201} source was always compared with the counting rate of a Bi^{207} source. Since the half-life of Bi^{207} is long (8.0 y), no corrections due to its decay need be applied. In Fig. 1 the ratio of the two counting rates, both corrected for background, is plotted in a semi-logarithmic diagram. Due to small impurities in the source, the curve is found to deviate slightly from a straight line. Since the measurements were started about one weak after the source preparation, the influence from the activities having shorter half-lives than Tl^{201} was estimated to be very small and quite negligible after a few days. Then the only impurity which could influence the measurements should be Tl^{202}, having a half-life of about 12 days. It was found that even small admixtures of this isotope become important at the end of the curve. The maximum contribution of Tl^{202} was calculated to be approximately 0.3 per cent at the beginning of the measurement. Taking this admixture into account, the following value of the half-life was obtained:

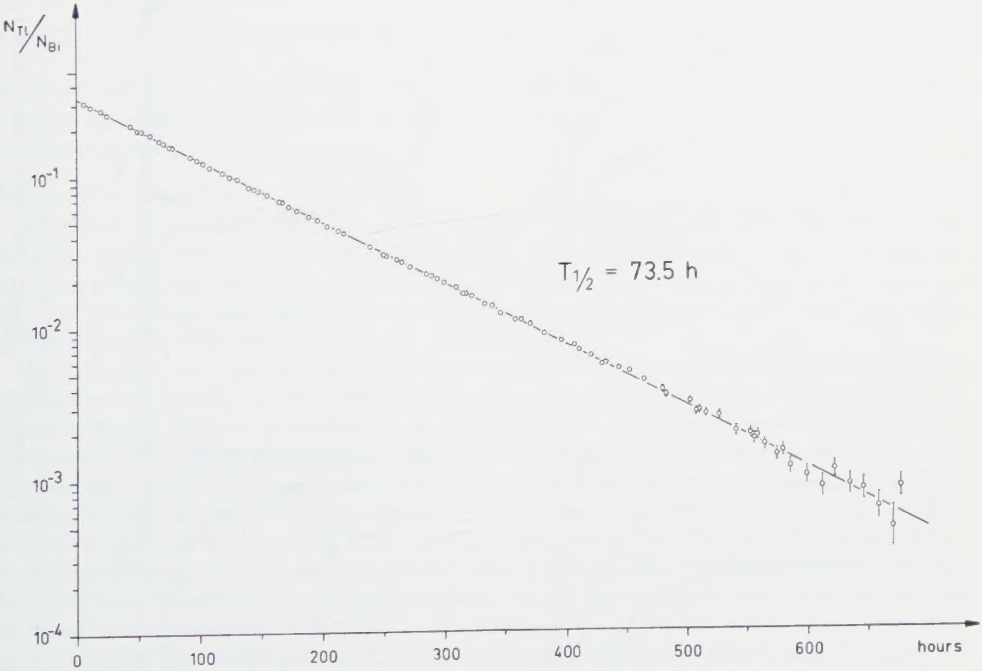

Fig. 1. Half-life of Tl^{201} obtained by use of an isotope-separated sample. The counting rate, measured with a G.M. counter, was determined in relation to the counting rate of a Bi^{207} source.

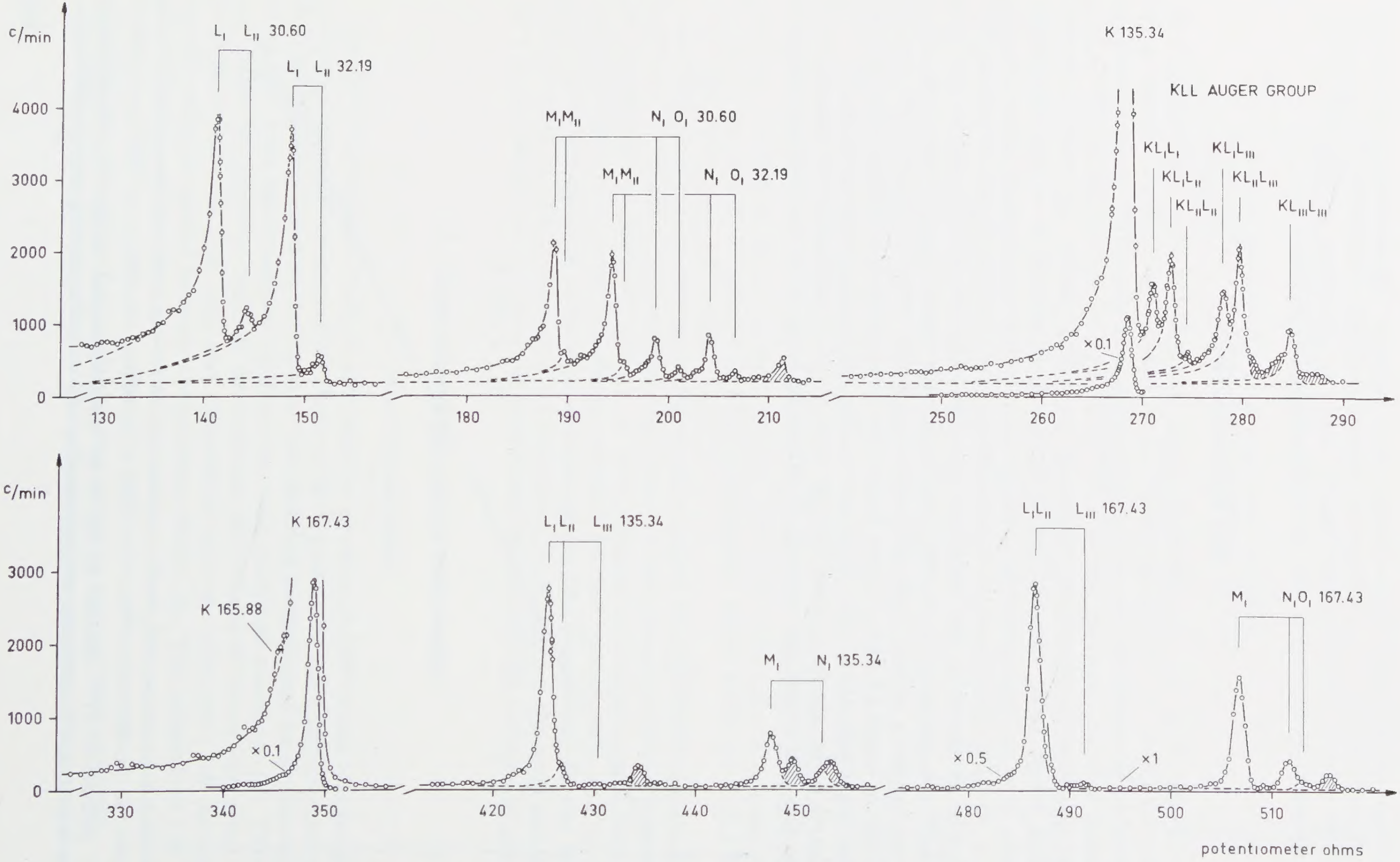

Fig. 2. The principal parts of the internal conversion spectrum measured in a double-focusing beta spectrometer. The spectrum is not corrected for decay. The dashed areas were found to originate from short-lived isotopes.

$$T_{1/2}(\mathrm{Tl}^{201}) = (73.5 \pm 0.8)\,h,$$

which is well within the limits of the value given by Neumann and Perlman: (72 ± 3) h [1].

3. Internal conversion measurements

The conversion measurements were carried out in a double-focusing beta spectrometer [7]. The window of the G.M. tube consisted of a 100 μg/cm^2 formvar foil supported on a brass grid. From the cut-off energy, measured to be (5.0 ± 0.5) keV, it can be concluded [8] that intensity corrections due to absorption in the window had to be applied only to the electron lines of the very lowest energy (i.e. about 15 keV), in which case the transmission factor comes out to be 99 per cent.

a. Energies and intensities

Fig. 2 shows the energy range 15–170 keV of the electron spectrum obtained with the spectrometer adjusted to a relative half-width of about 0.3 per cent. The energy region 170–640 keV was also investigated, but no Tl^{201} line with an intensity larger than about 0.1 per cent of the K 167.43 line intensity was detected here. Thus the conversion lines corresponding to the 0.55 MeV gamma ray emitted in the decay of Au^{201} [3] were not observed.

The half-lives of the various conversion lines were checked by repeating the measurements six days later. The energy calibration was performed 1) by use of the measured KLL Auger lines and the energy values deduced from the experimental data of Bergström and Hill [9], and 2) by the requirement of consistency between the K-, L_{I}- and M_{I}-conversion lines of the 135.34 and 167.43 keV transitions. The energy values thus obtained are given in Table 1 together with the intensity values.

As will be described below, a special run was made at higher resolution with an electromagnetically isotope-separated sample. In this case, as is clear from Table 1, the energies were obtained by the aid of energy values determined in the above-mentioned calibration.

b. The new transition of 165.88 keV

From the level scheme suggested by Bergkvist *et al.* [4] an E2 transition is expected to take place between the 1/2 − (167.2 keV) and 5/2 − (1.5 keV) states. However, no such transition was detected by Bergkvist *et al.*, which was supposed to be due to its low transition and conversion probabilities in comparison with the competing $M1$ transitions from the same level.

In the first run (at 0.3 per cent resolution) of the present investigation, weak indications of peaks were found which could be interpreted as the K-, L_{I}-, M_{I}- and N_{I}-conversion lines of a transition of about 166 keV. However, because of their low intensities and their positions on the low energy tails of the corresponding lines of the strong 167.43 keV transition, no definite conclusions could be drawn from this measurement. It was therefore decided to prepare a thinner source using electromagnetic isotope separation. With a sample thus obtained, the actual regions of the conversion-line spectrum was measured with 0.17 per cent resolution in the double-focusing spectrometer. As can be seen from Fig. 3, the statistics and resolving power in this measurement are such that the existence of a 165.88 keV transition, converted in Hg, can be definitely proved from the measured energies of the K and L_{I} lines (see Table 1). Furthermore, from repeated measurements, the lines were found to decay with the Tl^{201} half-life.

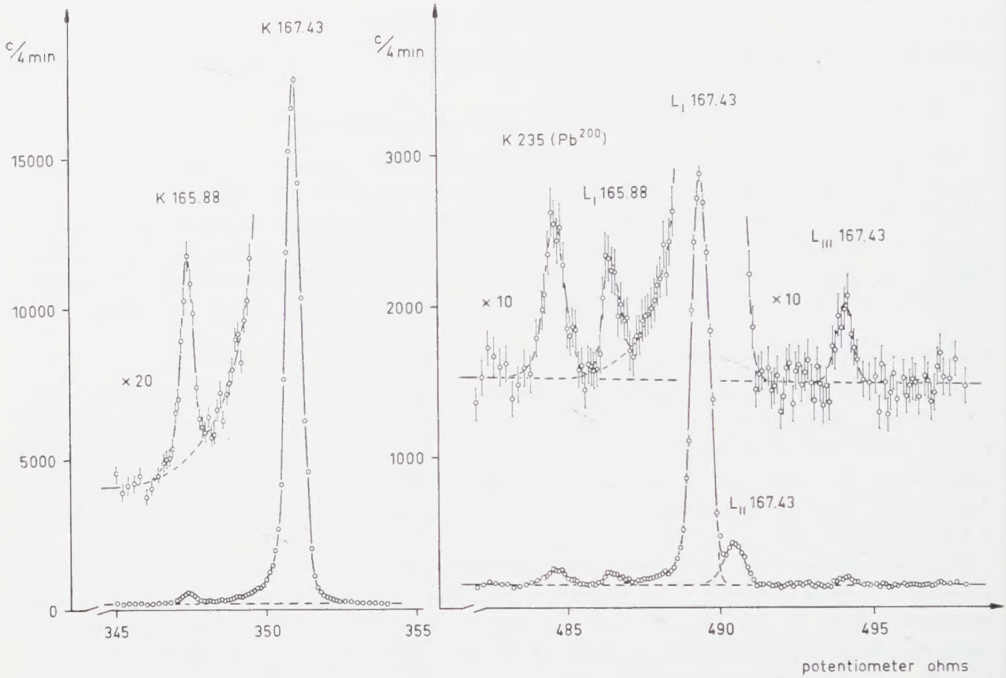

Fig. 3. Identification of the 165.88 keV transition from its weak internal K- and L-conversion lines situated on the low-energy tails of the strong lines of the 167.43 keV transition. Resolution: 0.17 %. The source was prepared by means of electromagnetic isotope separation. No decay corrections have been made.

The energy difference between the 167.43 and 165.88 keV transitions is thus found to be 1.55 ± 0.03 keV. This value should be compared with the energy difference 1.59 ± 0.02 keV obtained from the L-conversion lines of the 32.19 and 30.60 keV transitions, which were also studied with the isotope-separated sample (see Fig. 4). The 165.88 keV transition will be further discussed in the following section (see also Section C.1).

c. *Conversion ratios and multipolarity assignments*

The experimental information about internal conversion ratios of the five transitions found in the decay of Tl^{201} is summarized in Table 2. For the 135.34 and 167.43 keV transitions the K-conversion coefficients are also given, which were obtained from the external conversion measurements described below (see Section A.4). In cases where theoretical values are available, these have been included, for the sake of multipolarity determination, for four different multipolarities, $M1$, $M2$, $E1$ and $E2$. These values have been found from the tables of K- and L-conversion coefficients according to Sliv and Band [10] and from Rose's table of M-conversion coefficients [11]. However, theoretical $\Sigma L/\Sigma M$ ratios calculated with the available coefficients (L-conversion coefficients corrected for screening and for the finite size of the nucleus [10, 11], M-conversion coefficients *not* corrected for these effects [11]) turn out to be considerably lower (about 40 per cent) than what is generally found experimen-

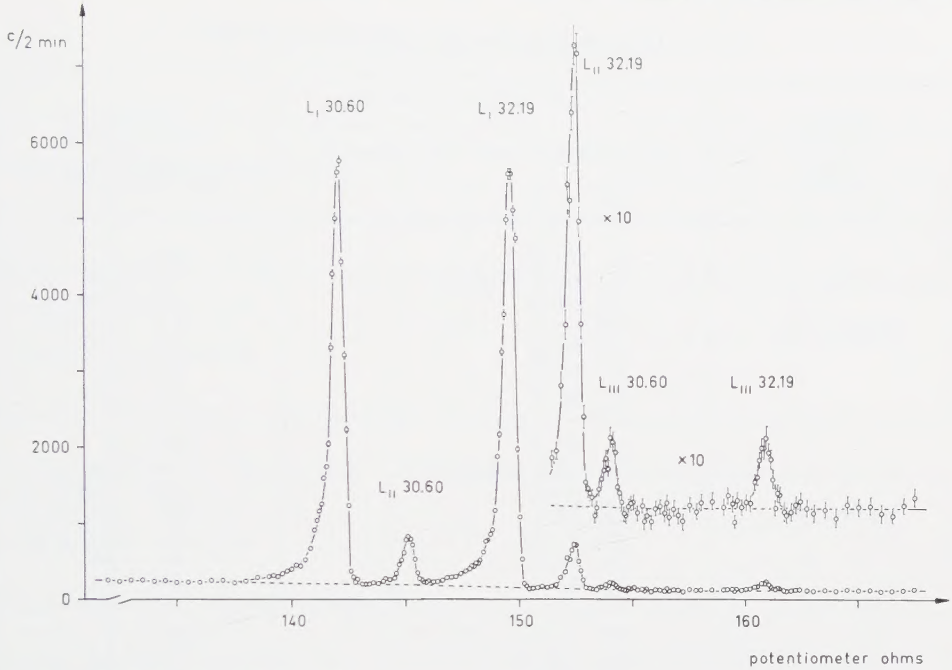

Fig. 4. Internal L-conversion lines of the 30.60 and 32.19 keV transitions, as measured with about 0.45 % resolution in a double-focusing beta spectrometer, using an isotope-separated sample. The lines have not been corrected for decay.

tally and might therefore be doubtful.[1] For this reason they have been omitted in Table 2.

As shown in the table, the multipolarity $M1$ can be unambiguously assigned to each of *the transitions of 30.60, 32.19, 135.34 and 167.43 keV*. For the two first-mentioned transitions, the $L_I/L_{II}/L_{III}$ ratios, as well as the M_I/M_{II} ratios suggest this assignment, and for the two latter transitions the experimental data of $K/\Sigma L$, $L_I/L_{II}/L_{III}$ and ε_K are all consistent with the theoretical values for $M1$. Furthermore, the fact that the M-conversion lines of the four transitions were found to originate only from the lowest M shells (cf. Table 1) agrees with Rose's theoretical conversion coefficients [11] for magnetic dipole radiation. For any other multipolarity, conversion electrons from higher M shells (especially M_{III}) should have been observed. The result of these multipolarity determinations is thus in agreement with that obtained by Bergkvist *et al.* [4].

The new transition of 165.88 keV can, on the basis of $K/L_I = 6.6 \pm 1.5$, be assigned $M1$ or $E2$, since the theoretical values [10] are 6.4 and 7.6, respectively. However, from the experimental limits of the L_{II}- and L_{III}-line intensities ($L_I/L_{II} > 6$ and $L_I/L_{III} > 7$), it can be concluded that the transition is mainly of multipolarity $M1$ (cf. the theoretical values [10]: 9.7 ($M1$), 0.17 ($E2$) for L_I/L_{II}, and 114 ($M1$), 0.25 ($E2$) for L_I/L_{III}). The maximum admixture of $E2$ radiation is found to be 7 per cent. As mentioned above, Bergkvist *et al.* [4] assumed multipolarity $E2$ for this transition, since it was undetected in their measurements.

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[1] Cf. e.g. C. de Vries, thesis (in print), Inst. v. Kernphysisch Onderzoek, Amsterdam.

Table 1. Gamma transitions in the decay of Tl^{201}.

Adopted transition energy (keV)	Internal conversion lines measured	Relative conversion-line intensity (K 167.43 = 100)
30.60 ± 0.03	L_{I} 30.59	50.9 ± 4
	L_{II} 30.61[1], 30.63	5.0 ± 0.6
	L_{III} 30.61[1]	0.56 ± 0.08
	M_{I} 30.61	14.2 ± 1.5
	M_{II} 30.60	1.5 ± 0.5
	N 30.61	4.0 ± 0.5
	O_{I} 30.59	0.7 ± 0.15
32.19 ± 0.03	L_{I} 32.19	50.2 ± 4
	L_{II} 32.19[1], 32.19	4.7 ± 0.6
	L_{III} 32.2[1]	0.65 ± 0.10
	M_{I} 32.17	12.6 ± 1.3
	M_{II} 32.23	1.5 ± 0.5
	N_{I} 32.17	3.1 ± 0.4
	O_{I} 32.19	0.6 ± 0.1
135.34 ± 0.04	K 135.34	56.0 ± 4
	L_{I} 135.34	7.9 ± 0.7
	L_{II} 135.37	0.77 ± 0.15
	L_{III} 135.38	0.07 ± 0.03
	M_{I} 135.38	2.2 ± 0.3
	N_{I} 135.30	0.60 ± 0.09
165.88 ± 0.07[2]	K 165.88[2], 165.90[2], 165.91[2]	1.65 ± 0.20
	L_{I} 165.86[2], 165.86[2]	0.25 ± 0.05
	M_{I} 165.8 ?	
	N_{I} 165.9 ?	$L_{\mathrm{II}} < 0.05$; $L_{\mathrm{III}} < 0.04$
167.43 ± 0.07	K 167.43	100
	L_{I} 167.45	14.6 ± 1.2
	L_{II} 167.41[2], 167.42[2]	1.6 ± 0.2
	L_{III} 167.59, 167.42[2], 167.42[2]	0.18 ± 0.04
	M_{I} 167.39	4.0 ± 0.4
	N_{I} 167.36	1.1 ± 0.15
	O_{I} 167.4	0.27 ± 0.06

[1] Energy measured in relation to L_{I} 30.60 and L_{I} 32.19.

[2] Energy measured in relation to the 167.43 keV energy. Relative error = ±0.03 keV.

After the assignment of multipolarities to the transitions, it is possible to obtain gamma-ray intensities and total transition intensities. These are calculated using the theoretical conversion coefficients [10] and the experimental conversion intensities given in Table 1. The resulting values are given in Table 3, columns 3 and 6. For comparison, the table includes also the gamma-ray intensities found from the external conversion measurements (Section A.4) and from the scintillation measurements (section A.5).

Table 2. Deduction of multipolarities.

ran- tion ergy eV)	Internal conversion relations						Multipolarity suggested[2]
	Quantity	Experimental values	Theoretical values[1]				
			$M1$	$M2$	$E1$	$E2$	
0.60	$L_I/L_{II}/L_{III}$	$100/(9.8 \pm 0.8)/(1.1 \pm 0.15)$	100/10.2/1.05	100/6.05/51.1	100/99/134	100/4800/5400	$M1$
	$\Sigma L/\Sigma M$	3.6 ± 0.4					
	M_I/M_{II}	9.7 ± 3	9.4	13	1.20	0.023	
	$\Sigma M/\Sigma N$	3.9 ± 0.5					
2.19	$L_I/L_{II}/L_{III}$	$100/(9.4 \pm 0.8)/(1.3 \pm 0.2)$	100/10.4/1.06	100/6.17/49.4	100/93/124	100/5000/5400	$M1$
	$\Sigma L/\Sigma M$	3.9 ± 0.4					
	M_I/M_{II}	8.4 ± 3	9.4	13	1.23	0.023	
	$\Sigma M/\Sigma N$	4.6 ± 0.6					
5.34	$K/\Sigma L$	6.4 ± 0.6	5.74	2.93	5.38	0.42	$M1$
	$L_I/L_{II}/L_{III}$	$100/(9.8 \pm 2)/(0.9 \pm 0.4)$	100/10.3/0.89	100/16.0/21.6	100/32/30	100/930/690	
	$\Sigma L/\Sigma M$	4.0 ± 0.5					
	$\Sigma M/\Sigma N$	3.7 ± 0.4					
	ε_K	$3.1^{+0.9}_{-0.6}$	2.77	15.3	0.16	0.40	
5.88	K/L_I	6.6 ± 1.5	6.4	4.1	8.5	7.6	$M1$[3]
	L_I/L_{II}	>6	9.7	7.0	3.6	0.17	
	L_I/L_{III}	>7	114	6.7	3.8	0.25	
7.43	$K/\Sigma L$	6.2 ± 0.5	5.74	3.25	5.56	0.69	$M1$
	$L_I/L_{II}/L_{III}$	$100/(11.0 \pm 1.5)/(1.2 \pm 0.3)$	100/10.2/0.88	100/14.3/14.9	100/28/26	100/600/400	
	$\Sigma L/\Sigma M$	4.1 ± 0.5					
	$\Sigma M/\Sigma N$	3.6 ± 0.5					
	ε_K	$1.5^{+0.4}_{-0.3}$	1.50	7.2	0.096	0.26	

[1] $K/\Sigma L$ values, L-subshell ratios and ε_K values according to Sliv and Band [10]; M-subshell ratios from the table of Rose [11]. Theoretical $\Sigma L/\Sigma M$ values have been omitted for the reason given in the text (see Section A.3.c).

[2] The fact that M conversion is observed only in the lower shells (cf. Table 1) also confirms the magnetic dipole assignments. According to Rose's theoretical M-conversion coefficients [11] higher M lines should have been detectable for any other multipolarity.

[3] The experiments do not exclude an $E2$ admixture of up to 7 %.

4. *External conversion measurements*

As mentioned in the preceding section, absolute values of the internal K-conversion coefficients were determined for the transitions of 135.34 and 167.43 keV. Following the method described by Hultberg and Stockendal [12, 13], internal and external conversion lines were measured with the same source (2×12 mm^2 in our case) and with the same slit setting in the double-focusing spectrometer.

The *external* conversion measurements were carried out with a 2.19 mg/cm^2 uranium converter (13.5×20 mm^2, "razor-blade" shape [13]) and by use of 0.4 mm aluminium for the absorption of internal conversion electrons. The spectrum thus obtained is shown in Fig. 5 (the low-energetic K lines were not measured).

The *internal* conversion lines measured (M, N, O group of the 167.43 keV transition) are shown in the inset of Fig. 5. The K intensities of interest for the calculations below were inconveniently large for the G.M. tube, but could be deduced by the aid of the relative intensity values of Table 1.

Following the notation in ref. [12], we may write:

$$\varepsilon_K = \frac{A_{\beta_K}}{A_{\gamma_L}} \tau_L \, f_L \, d \, b \, ,$$

where

ε_K = internal K-conversion coefficient
A_{β_K} = recorded intensity of internal K-conversion electrons
A_{γ_L} = recorded intensity of L photoelectrons
τ_L = photoelectric cross-section for the L shell (see below)
f_L = correction factor for τ_L (see ref. [12] and below)
d = converter thickness
b = dimension factor [12].

The τ_L values were obtained from the tabulated values of τ_K [14] by use of the relation $\tau_K/\tau_L = 5.3 \pm 0.2$ [13].

As regards f_L, interpolated values could be found by the aid of the longitudinal angular photoelectric distributions $J(\theta)$ at the gamma-ray energies 412, 662 and 1332 keV [13], and from the value at the K-threshold energy. The latter value was calculated using the relation $J(\theta) = \sin^2 \theta$ [12], since $f_L \rightarrow f_K$ at low energies (e.g. $f_K/f_L = 1.005$ at 412 keV). However, since there are no experimental distributions available below 412 keV, the f_L values thus obtained for 135.34 and 167.43 keV are somewhat uncertain. The errors have been estimated as $^{+25}_{-10}$ % and $^{+15}_{-10}$ %, respectively.

Inserting numerical values for the quantities in the above formula for ε_K, one obtains:

$$\varepsilon_{K135.34} = 3.1^{+0.9}_{-0.6}$$

$$\varepsilon_{(K165.88 + K167.43)} = 1.5^{+0.4}_{-0.3} ,$$

where the errors given have been obtained by quadratic addition of the separately estimated uncertainties, namely for A_{β_K} 7 %, A_{γ_L} 15 %, τ_L 7 % [12, 13] and d 1 % [12], (error in f_L, see above). The contribution due to the uncertainty in the half-life could be neglected. Slight corrections (of the order of 1 %) were made for the gamma-ray absorption in the aluminium and for the extension of the source.

The theoretical values of the K-conversion coefficient [10] are, for the multipolarity $M1$, 2.77 at 135.34 keV and 1.50 at 167.43 keV, in good agreement with the experimental values. Since the theoretical values for any other multipolarity are far outside the experimental limits (see Table 2), it can be concluded that the 135.34 and 167.43

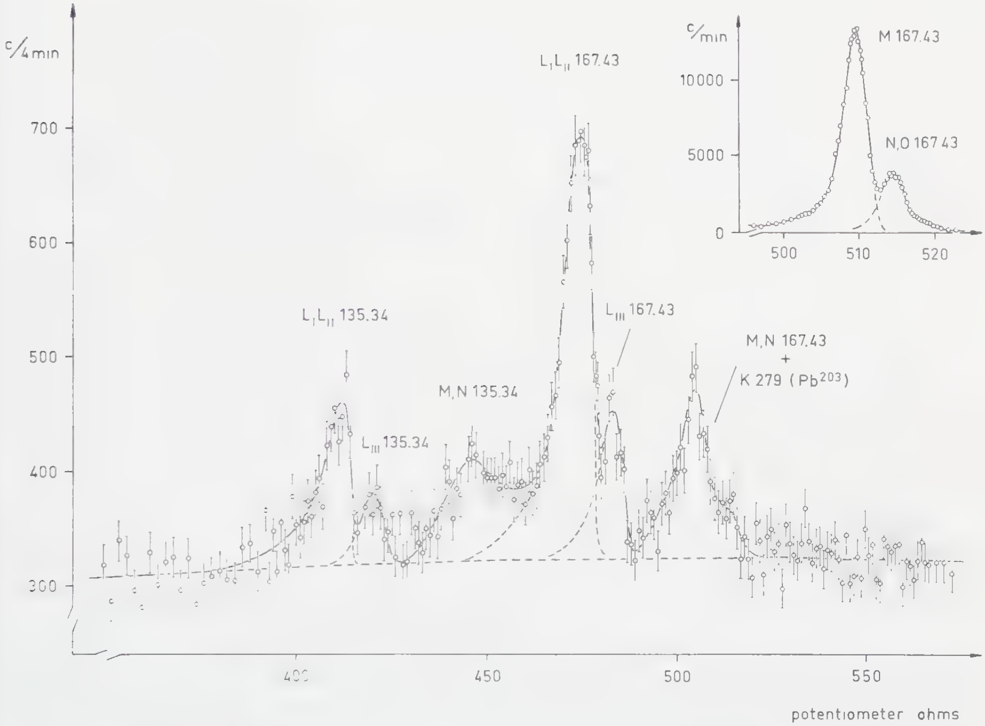

Fig. 5. External conversion lines of the 135.34 and 167.43 keV transitions, as measured with a resolution of about 1.2 % in a double-focusing beta spectrometer. A 2.19 mg/cm² U converter was used. For the absolute determination of internal conversion coefficients, the internal conversion lines shown in the inset were measured with the same source and with the same slit setting. The lines are not corrected for decay.

keV transitions both have mainly magnetic dipole character.[1] As pointed out in the preceding section, also the measured internal conversion ratios lead to the same conclusion (see also Table 2).

As additional information the external conversion measurements also give an estimate of relative probabilities for the photoeffect in the *L* subshells of uranium at the energies 135 and 167 keV (see Appendix 2).

5. *Scintillation measurements*

The gamma-ray spectrum of Tl^{201} was studied in a scintillation spectrometer with a 1″ × 1½″ NaI(Tl) Harshaw crystal. The pulse-height spectrum was analysed in a multi-channel analyser of the Hutchinson-Scarrott type. These measurements were closely connected to a determination of the electron-capture decay energies of Tl^{201} and Tl^{202} and are presented in a separate paper by Gupta [15]; (the scintillation spectrum is shown in Fig. 1. ref. [15]). The relative gamma-ray intensities obtained are given below in Table 3, column 5.

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[1] Regarding the weak transition of 165.88 keV, no conclusions can be drawn from the external conversion measurements.

Table 3. Gamma-ray intensities and total transition intensities in the decay of Tl^{201}.

Transition energy (keV)	Multipolarity	Gamma-ray intensity (K 167.43 = 100)			Total transition intensity[2]
		From internal conversion spectrum[1]	From external conversion spectrum	From scintillation spectrum	
30.60	$M1$	1.5 ± 0.1		< 7	78 ± 5
32.19	$M1$	1.8 ± 0.1			75 ± 5
135.34	$M1$	20 ± 1.5	18 ± 4	22 ± 2	88 ± 4
165.88	$M1$[3]	1.1 ± 0.15	68	68	3.1 ± 0.3
167.43	$M1$	67			188
K X-rays				725 ± 40	

[1] Values calculated by use of the K- (or L-) conversion-line intensities, given in Table 1, and the theoretical conversion coefficients [10].

[2] Values obtained from the gamma intensities in column 3, this table, and the experimental conversion intensities from Table 1.

[3] The possible $E2$ admixture ($< 7\%$) will leave the given intensity values unchanged.

The intensities of the 30.60 and 32.19 keV gamma rays cannot be determined directly from the scintillation spectrum, since the escape peak of the strong K X-ray peak falls in the same region. An upper limit of the total intensity of these two lines has been estimated from the difference of two Tl^{201} spectra measured with and without a copper absorber of 160 mg/cm^2.

Due to an admixture of Tl^{200} and Tl^{202}, which gave small contributions to the Tl^{201} gamma lines, a correction varying between 1 and 2 per cent had to be applied.

6. *Coincidence measurements*

Electron-gamma coincidence measurements were performed in an intermediate-image beta spectrometer [16] fitted with a $1'' \times 1\frac{1}{2}''$ NaI(Tl) crystal for the detection of the gamma rays. The beta detector consisted of a G.M. tube with a 100 μg/cm^2 formvar window for the detection of the low-energy electrons. In order to get a good energy resolution in the scintillation channel, the crystal was mounted directly onto the photomultiplier. The system was then placed inside the pole-piece of the spectrometer. To compensate for the magnetic stray-field, a weak current was sent through a coil wound around the photomultiplier. This arrangement, as well as the coincidence circuit used, is described in detail by Johansson [17]. The resolving time (2τ) of the coincidence set-up was about 0.2 μsec.

In the two beta-gamma coincidence experiments carried out, the spectrometer current was varied over the partly resolved L lines from the low-energy transitions of 30.60 and 32.19 keV. The coincidence curve shown in Fig. 6 was obtained with the 135.34 keV gamma rays entering the scintillation channel. As a weak, electromagnetically separated source was used, the rate of accidental coincidences was very low (about 1 per cent of the genuine coincidence rate). It can thus be concluded from the figure that the 30.60 and 32.19 keV transitions are both in coincidence with the 135.34 keV transition.

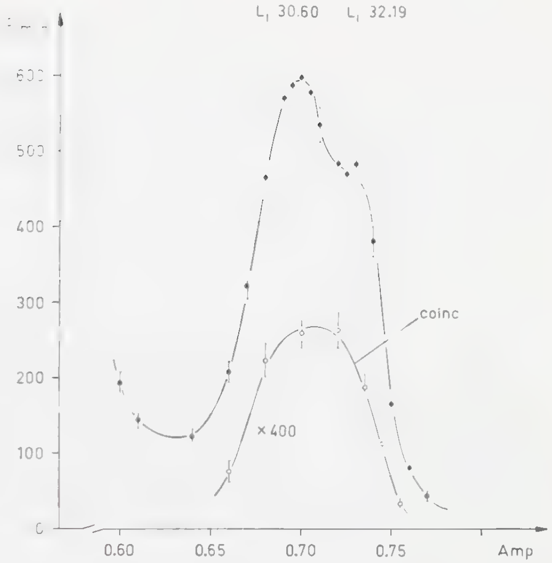

Fig. 6. Electron-gamma coincidences (lower curve) between the (L 30.60 + L 32.19) group and the 135.34 keV gamma ray. The measurements were carried out by use of an intermediate-image beta spectrometer. The upper curve shows the counting rate in the beta channel. The accidental coincidence rate was negligible. Decay corrections have been performed.

When the 167.43 keV gamma ray was selected in the scintillation channel, no coincidences were observed between this transition and the two low-energy L lines. In another experiment, using scintillation detectors in both channels, it was shown that also the gamma ray of 135.34 keV is not in coincidence with the one of 167.43 keV.

It can thus be concluded that the above results are in agreement with those obtained by Bergkvist *et al.* [4]. No coincidence experiments were performed with the 165.88 keV transition because of the difficulties in resolving this weak transition from the much stronger one of 167.43 keV.

B. Construction of the decay scheme

According to the present investigation, the most probable decay scheme of Tl^{201} is the one presented in Fig. 7. It is gratifying to find that the level sequence is in agreement with that proposed earlier by Bergkvist *et al.* [4].

1. Level sequence

Due to the accuracy in the energy measurements (see Table 1), the following energy relations strongly suggest that the transitions of 165.88 and 167.43 keV are cross-over transitions:

$$30.60+135.34=165.94 \text{ (cf. transition energy}=165.88 \text{ keV)}$$
$$32.19+135.34=167.53 \text{ (cf. transition energy}=167.43 \text{ keV)}$$

The results of the coincidence measurements also lend strong support to this suggestion. Since the 167.43 keV transition was found not to be in coincidence with any other transition, it seems quite natural to assume that the 167.43 keV transition de-excites a state (the highest one) of the same energy. The coincidences observed between the 135.34 keV gamma ray and the two low-energy transitions, in combination with the intensity relation $I_{tot}(30.60+32.19) > I_{tot}(135.34)$ (see Table 3, column 6), then unambiguously propose the positions of the intermediate levels shown in Fig. 7.

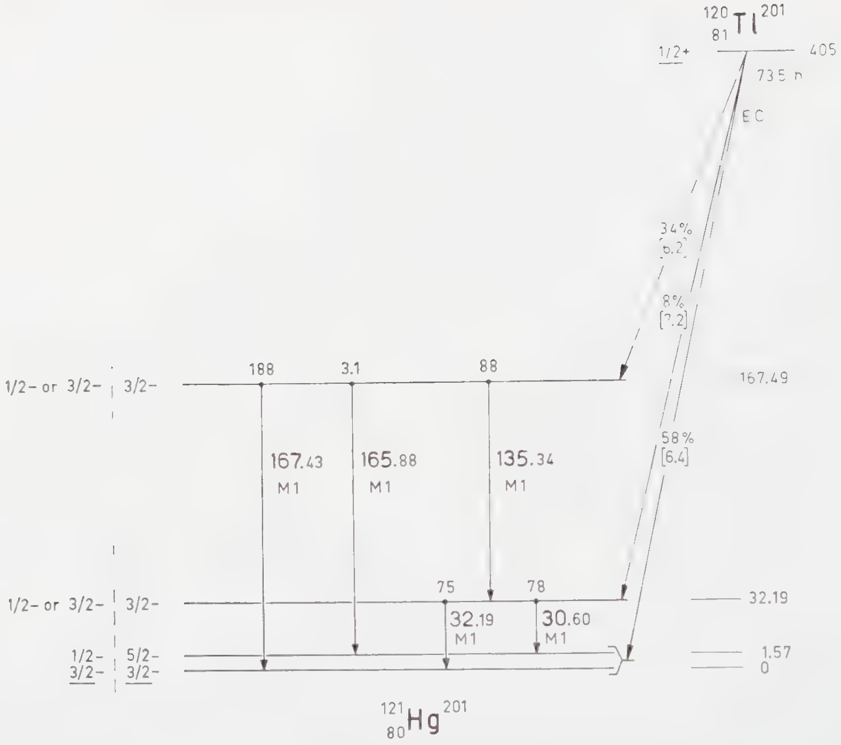

Fig. 7. Decay scheme of Tl^{201}. The different possibilities for spin assignments are discussed in the text. The total transition intensities, indicated at the head of the transitions, are given in the intensity scale used in Tables 1 and 3 (*K* 167.43 = 100). Because of the small level separations, the figure is not drawn to scale.

2. Level energies

Due to the possibility of obtaining the level energies from different combinations of the measured energy values of the transitions, one finds slightly different values for the level energies. The values given in Fig. 7 have then been chosen as weighted mean values. Thus, the energy of the 1.57 keV level was obtained by considering the differences between the transition energies 167.43 and 165.88 keV, and 32.19 and 30.60 keV, respectively. In the same way the energy of the highest level was calculated as a weighted mean value of 167.43 keV and (32.19 + 135.34) keV.

3. Spin assignments

So far the present investigation has mainly given results in agreement with those obtained by Bergkvist *et al.* [4]. However, because they did not observe the 165.88 keV transition between the 167.49 and 1.57 keV levels, they suggested a spin difference of two units between these states. The unobserved gamma transition should then be of multipolarity $E2$ and therefore have much lower transition and conversion probabilities than the competing $M1$ transition to the ground state. Assuming an $s_{1/2}$ ground state in Tl^{201}, the following level assignment for Hg^{201} agreed with their experiments: $p_{3/2}$ (0 keV), 5/2 − (1.5 keV), 3/2 − (32.1 keV), and 1/2 − (167.2 keV). The ground-state spin of Hg^{201} was already known from spectroscopic measurements [18], and the spin value 1/2 of the Tl^{201} ground state has later been verified in magnetic resonance experiments [19].

Since the 165.88 keV transition now has been verified and has proved to be mainly of $M1$ character, the spin assignments above are partly in disagreement with experiments and have therefore to be reconsidered. It should be pointed out, however, that even the present knowledge of Hg^{201} is insufficient for definite conclusions to be made about the spin assignments of the excited states. Different possibilities of assignments are therefore given in Fig. 7 and will be discussed below.

Because of the $M1$ character of the transition from the 167.49 keV state to the 3/2 − ground state, the spin and parity of the first-mentioned state should be 1/2 −, 3/2 − or 5/2 −. However, according to the following consideration, the 5/2 − assignment seems impossible. In order to determine the total decay energy of Tl^{201}, Gupta [15] measured the fraction of the electron-capture transitions to the 167.49 keV state taking place by K capture. Then, by assuming that the 167.49 keV state is 1/2 − or 3/2 − (non-unique, first forbidden transition), the decay energy was calculated using the theoretical results of Brysk and Rose [20]. By the aid of the additional information about electron-capture branching ratios (see next section), the log ft value of the K-capture transition to the 167.49 keV level can be calculated to be about 6, which is thus consistent with the assumption of a non-unique, first forbidden transition. On the other hand, if the corresponding calculations are carried out on the assumption of a 5/2 − assignment of the 167.49 keV state (corresponding to a unique, first forbidden transition), one obtains a log ft value of about 6.5. This is in disagreement with the assumption made, which would require a log ft value of about 9. In conclusion, therefore, the 167.49 keV state most probably should be assigned either 1/2 − or 3/2 −.

In a similar way spin and parity can be proposed for the 32.19 keV state. Since it is populated by an $M1$ transition from the 1/2 − or 3/2 − state of 167.49 keV, and also since it is de-excited to the 3/2 − ground state by an $M1$ transition, the 32.19 keV state should be assigned 1/2 −, 3/2 − or 5/2 −. Also in this case, however, the 5/2 − assignment might be excluded, since the log ft value of the K-electron capture transition to the 32.19 keV state is found to be about 7, indicating a non-unique, first forbidden transition (cf. next section).

The assignment of the 1.57 keV state is more complicated. Since it is populated by $M1$ transitions from both of the higher levels, its spin and parity might be 1/2 −, 3/2 − or 5/2 −. A few remarks can be made on the choice. The assignment 3/2 − seems least probable, since it would give rise to a strong configuration interaction with the ground state, resulting in a level splitting larger than experimentally found [21]. It should further be pointed out that the spin value 5/2 can be combined only with the spin values 3/2 of the higher states, but is incompatible with the alternative suggestions of 1/2. It may be commented here that an assignment 5/2 − to the first excited state would give a reasonable explanation of the low transition probability of the 165.88 keV transition (cf. Table 3 and Section C.1), since the latter, in this case, should be l-forbidden. On the other hand, this also implies the 30.60 keV transition to be l-forbidden, which is not indicated in the experiments.

To sum up, therefore, the following spin and parity assignments are proposed for the states in Hg^{201}:

3/2 − (0 keV), 5/2 − (1.57 keV), 3/2 − (32.19 keV), and 3/2 − (167.49 keV);

or

3/2 − (0 keV), 1/2 − (1.57 keV), 1/2 − or 3/2 − (32.19 keV),
and 1/2 − or 3/2 − (167.49 keV).

4. *Electron-capture feeding*

In Fig. 7 the branching ratios in the electron-capture decay of Tl^{201} to the different levels in Hg^{201} are given in per cent per disintegration. For the two lowest levels only the summed electron-capture feeding could be obtained, since the number of transitions taking place between these levels is unknown. In the calculation of the branching ratios only non-unique, first forbidden transitions have been considered. This limitation, however, is justified as will be shown in the following brief description of the procedure of the calculations.

The relative electron-capture intensities to the two highest levels are obtained directly from the transition intensities, given in Table 3, column 6. The feeding to the two lower levels, however, has to be calculated in a more complicated way. Since the fraction of the electron-capture transitions to the highest state taking place by K capture, P_K, has been measured [15], one can find the number of K-capture transitions to this state. Furthermore, since the total decay energy of Tl^{201} is known, 405^{+70}_{-40} keV [15], the P_K value for the electron-capture transitions to the 32.19 keV state can be found from the theoretical results of Brysk and Rose [20], thus giving the intensity of the K-capture transitions to this state. The total K-capture intensity of the transitions to the two lowest states is then found by subtraction from the total K-capture intensity in the decay of Tl^{201}. The latter intensity is obtained from the measured K X-ray intensity (see Table 3, column 5) after correction for fluorescence yield and conversion in the K shell. Finally, using a theoretical P_K value as above, one finds the total electron-capture feeding to these states. The resulting branching ratios are in per cent per disintegration (level energies given in parentheses): 34 ± 4 (167.49 keV), 8 ± 2 (32.19 keV) and 58 ± 5 (1.57 keV + ground state). The log ft values for the K-electron capture transitions to these levels are 6.2, 7.2 and 6.4, respectively.

As pointed out above, the calculations were carried out on the assumption of non-unique, first forbidden transitions. This assumption is self-evident in the case of the total feeding to the two lowest states, since the ground state is assigned $3/2-$. In the case of the other two electron-capture transitions, the assumption is justified according to a consideration of log ft values. If the total decay energy, P_K values and branching ratios are calculated by assuming that one or both of the upper states are assigned $5/2-$ (corresponding to unique, first forbidden transitions), the resulting log ft values are too low (≈ 7.5) to be consistent with the assumption made (cf. the preceding section and ref. [15]).

C. Discussion

1. *Gamma-transition probabilities*

Since all the four strong transitions have been classified as almost pure $M1$ transitions, it might be interesting to compare the experimental intensity ratio for the gamma rays de-exciting a level with the ratio obtained from the theory of Weisskopf for single-proton transitions [22]. From the gamma-intensity values given in Table 3, column 3, one finds for the 167.43 and 135.34 keV gamma rays the ratio $I_{\gamma 167.43}/I_{\gamma 135.34} = 3.3$, while the corresponding theoretical value is found to be 1.9. For the two low-energy gamma rays, both experiment and theory give $I_{\gamma 32.19}/I_{\gamma 30.60} = 1.2$. So far the experimental values are found to be in good agreement with theory. It is therefore interesting to find the low intensity ratio for the 165.88 and 167.43 keV gamma transitions. From the experimental data (see Table 2), the 165.88 keV transition is proposed to have mainly magnetic dipole character. If it is regarded as a pure $M1$ transition, the experimental value of $I_{\gamma 165.88}/I_{\gamma 167.43}$ is found to be only about 1.7 per cent of the theoretical value. Thus, compared to both the 167.43 keV gamma transition and the one of 135.34 keV, the 165.88 keV transition is strongly hindered. According to this result, the possible $E2$ admixture mentioned in Section A.3.c seems quite feasible.

2. *Comparison with neighbouring isotopes*

In the neutron-deficient, even-odd isotopes in the neighbourhood of the double-magic Pb^{208}, $1/2-$, $3/2-$, $5/2-$, and/or $13/2+$ states have been found and interpreted as single-particle states according to the nuclear shell model. It is therefore tempting to assume that the states found in Hg^{201} (having five neutron holes) are of this type. If this be the case, it would be interesting to inspect how these levels fit into the systematic behaviour of the single-particle levels of, in the first place, the odd Hg isotopes. The levels experimentally found for these isotopes are shown in Fig. 8. However, since the information seems to be rather incomplete for some of the nuclei, and since no unambiguous assignments can be given to the excited states of Hg^{201}, an adequate comparison is very difficult to make. Hence only a few remarks can be made.

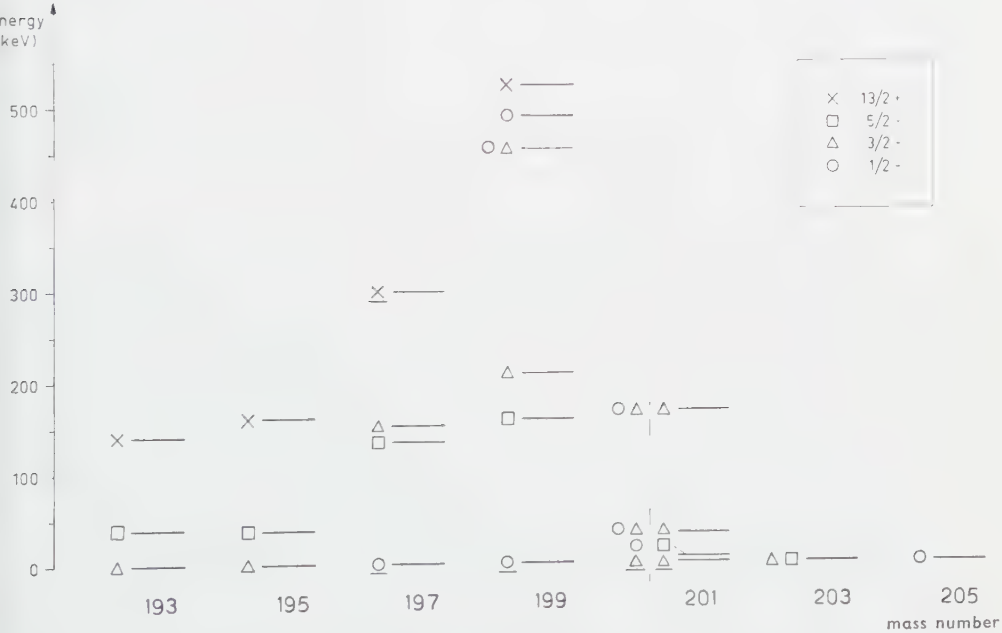

Fig. 8. Experimental energy levels in the odd Hg isotopes. The spin values of the underlined level assignments have been measured directly. An additional level in Hg^{201} may be located at 0.55 MeV, since a gamma ray of this energy is emitted in the beta decay of Au^{201} [3].

Starting with the 13/2 − and 5/2 − states, it is found that their energy separation (= energy of *M*4 transition) varies in a systematic way from Hg^{193} to Hg^{199} [23]. According to these systematics, an *M*4 transition of approximately 600 keV would be expected to be found in Hg^{201}. However, in an experiment performed by Bergström *et al.* [2], no isomeric transition was observed, which might indicate that its metastability has been destroyed by an underlying level of intermediate spin.

Also the energy separations between the 3/2 − and 5/2 − levels in Hg^{195} ,Hg^{197} and Hg^{199} lie on a smooth curve when plotted as a function of the odd neutron number [23]. From an extrapolation of this curve one might, in the case of Hg^{201}, expect a 3/2 − level situated 100–200 keV above a 5/2 − level. It is therefore interesting to find that one of the possible suggestions in Fig. 7 represents just such a case [3/2 − (167.49 keV), 5/2 − (1.57 keV)].

Some further features of the odd Hg isotopes may be pointed out (see Fig. 8). The ground states of Hg^{193} and Hg^{195} are assumed to be 3/2 − [24], whereas the spin values of the Hg^{197} and Hg^{199} ground states have been measured to be 1/2 [25, 18]. The spin value of 3/2 then reappears in Hg^{201} [18]. The ground state of Hg^{203} might be 3/2 − or 5/2 − [23, 26]. Finally, Hg^{205} probably has a 1/2 − ground state [23]. Thus it is seen that the ground-state spins show a great irregularity. Another anomaly is the sudden occurrence of very low-lying levels in Hg^{201}. Furthermore, as can be seen in Figs. 7 and 8, the experiments suggest that at least two of these low-lying states have the same spin and parity assignments, whereas the single-particle assignments in the neighbouring even-odd isotopes appear only once in each isotope.[1]

Thus one can summarize that the systematic consideration of the odd Hg isotopes reveals some clear regularities, but also, some large irregularities, the most striking of which seem to appear in Hg^{201} (cf. Fig. 8).[2] However, it must be kept in mind that this judgement might be erroneous due to the incompleteness of experimental information for some of these isotopes. Further experimental studies are therefore obviously called for in these cases, particularly with the purpose of clarifying the lowest energy regions of the level schemes. This recommendation includes measurements of the ground state spin values.

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[1] The only exceptional case known so far is Hg^{199}, where the assignments 1/2 − and 3/2 − at 0 keV and 208 keV, respectively, reappear in the higher states, 1/2 − (491 keV) and 1/2 − or 3/2 − (455 keV) [2]. These states, however, lie considerably outside the low energy region considered here.

[2] The occurrence of low-lying single-particle states in Hg^{201} having the same assignments might be understood in terms of the single-particle model [21]. Since the single-particle levels seem to lie rather close to each other, it would be possible, with only a small amount of energy, to lift a neutron pair from a level with lower spin up to a level with higher spin. The nuclear state arising from this pair excitation will have the same spin and parity as the original state. Furthermore, since the energy consumption might be partly compensated for due to the change in pairing energy, the two states might have not too different energies. Beyond the rough qualitative picture given here, theoretical predictions seem to be very difficult to make. Due to the large number of neutron holes, a quantitative calculation of the level energies in Hg^{201} would involve uncertainties of at least the same order as the level splittings experimentally found.

The systematics of the odd Hg isotopes might also be regarded from another point of view [27]. The unique properties of Hg^{201} might indicate a state intermediate between spherical and ellipsoidal equilibrium shapes. The slow oscillations of the "soft nucleus" may then give rise to the observed low-lying levels.

Appendix

1. Relative intensities of the KLL Auger lines in mercury

In a recent work by Asaad [28], theoretical, relativistic calculations of the positions and relative intensities of the *KLL* Auger lines in mercury are presented. The results of these calculations are found to be in agreement with the experimental values found by Bergström and Hill [9]. In their measurements, which were performed with a beta spectrometer with photographic recording, the stress was laid upon the accurate determination of the energies of the Auger lines. The intensities, on the other hand, are given with estimated errors of about 30 per cent. In the present investigation the *KLL* Auger lines were used for the energy calibration of the double-focusing spectrometer and therefore no comparison can be made between the experimental and theoretical energy values. The intensities, however, would be useful for a comparison with theory, since they could be more accurately estimated in the double-focusing spectrometer than in the spectrometer with photographic recording. It might be remarked that, as can be seen in Fig. 2, the Auger lines are not completely resolved in our measurements. However, since the line shape is accurately known from the well resolved *K* 135.34 line situated just below the $KL_{\mathrm{I}}L_{\mathrm{I}}$ Auger line, the intensity determinations are much facilitated.

Table 4 gives the intensity values, obtained in the present work, together with the theoretical values of Asaad [28] and the experimental values of Bergström and Hill [9]. A normalization of our intensity values to $KL_{\mathrm{I}}L_{\mathrm{I}} = 100$ would, for the five remaining lines, result in values which are all higher than the corresponding theoretical values. It was therefore considered more correct to present the results in a scale adjusted to Asaad's values by a weighted mean-square method. As can be seen from Table 4, the agreement between the experimental and theoretical values is rather good.

Table 4. Relative intensities of the *KLL* Auger lines in mercury.

Line assignment	Theoretical values according to Asaad	Experimental values	
		Bergström and Hill[1]	This work
$K\,L_{\mathrm{I}}\,L_{\mathrm{I}}$	100	100	88 ± 9
$K\,L_{\mathrm{I}}\,L_{\mathrm{II}}$	144	120	148 ± 11
$K\,L_{\mathrm{II}}\,L_{\mathrm{II}}$	9	20	12 ± 6
$K\,L_{\mathrm{I}}\,L_{\mathrm{III}}$	82	70	75 ± 9
$K\,L_{\mathrm{II}}\,L_{\mathrm{III}}$	146	140	162 ± 13
$K\,L_{\mathrm{III}}\,L_{\mathrm{III}}$	66	60	67 ± 7

[1] Intensity values given with estimated errors of about 30 per cent.

2. Relative photo cross-sections for L subshells of uranium

Since the L_{III} lines are resolved from the $(L_{\mathrm{I}} + L_{\mathrm{II}})$ lines in the external conversion spectrum shown in Fig. 5, it is possible to make an estimate of the ratio $(\tau_{L_{\mathrm{I}}} + \tau_{L_{\mathrm{II}}})/\tau_{L_{\mathrm{III}}}$ at the energies 135 and 167 keV, where the τ's indicate relative probabilities for the photoeffect in the L subshells of uranium. The values obtained from a determination of the areas under the photolines in Fig. 5 are given in Table 5.

Table 5. Experimental and theoretical values of $(\tau_{L_I}+\tau_{L_{II}})/\tau_{L_{III}}$ for $Z=92$.

$h\nu$ (keV)	Theoretical values	Experimental values
135	3.0	3.3 ± 0.6
167	3.6	3.9 ± 0.5

Recently a similar determination was performed by Grigor'ev and Zolotavin [29] using bismuth as converter for the gamma rays of energies 87, 121, 136 and 265 keV. Their experimental results were compared with theoretical values deduced by use of formulae of Stobbe [30], considering the ratio $\tau_{L_{II}}/\tau_{L_{III}}$ to be 0.45 for $Z=83$ [31, 32]. Following the same procedure, theoretical values of $(\tau_{L_I}+\tau_{L_{II}})/\tau_{L_{III}}$ can be found also in the present case ($Z=92$). By setting $\tau_{L_{II}}/\tau_{L_{III}}=0.43$ in accordance with the theory of Phillips [32], the Stobbe formulae [30] give the values shown in Table 5.

As can be seen from the table, the experimental and theoretical values coincide within the experimental errors. Although the errors are relatively large, it is interesting to note that the experimental values in this work, as well as those of Grigor'ev and Zolotavin (except for the highest energy) are, on the average, about 10 per cent higher than the theoretical values. However, when comparing the experimental and theoretical values, it must be kept in mind that the influence of the angular distributions of photoelectrons from the various L subshells have not been taken into account.

ACKNOWLEDGEMENTS

The authors would like to thank Dr. G. Andersson at the Gustaf Werner Institute for Nuclear Chemistry, Uppsala, for arranging the electromagnetic isotope separations. The irradiations performed at the same institute by Ing. Å. Svanheden and his cyclotron crew are gratefully acknowledged. Our thanks are also due to Prof. K. Bleuler and Dr. J. Blomqvist for their interest in the theoretical aspects of this work.

Finally we wish to thank Karin Oelsner, B.Sc., for correcting the English.

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Tryckt den 4 april 1960

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